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# Multicomponent Adsorption Isotherm Studies on Removal of Multi Heavy Metal Ions in MSW Leachate using Fly Ash

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***Abstract*** **- The present research work provides the investigation on simultaneous adsorption of multi heavy metal ions present in MSW leachate by lignite based Coal Fly ash. Municipal solid waste leachate samples were collected from the leachate pit, of Mayiladuthurai municipal solid waste open dumping site, Nagapattinam District, Tamil Nadu, India. Leachate samples were analyzed and multi heavy metal ions namely Cu, Zn, Pb, Cr and Ni were identified. The lignite based coal fly ash used for this study has been obtained from the Neyveli Lignite Corporation, a Public sector company located at a distance of 245km south of Chennai, the capital city of the state of Tamilnadu. Fly ash applicability for adsorption of such a multi heavy metal ions present in MSW leachate was studied by three Multicomponent adsorption isotherm models namely Extended Langmuir Isotherm model, Langmuir - Freundlich Isotherm model and Multicomponent Isotherm model. Experimental equilibrium metal uptake data were compared with calculated values. The performance of the model were studied based on the coefficient of correlation.**

***Key words: Fly Ash, Municipal solid waste leachate, Extended Langmuir Isotherm model, Langmuir - Freundlich Isotherm model, Multicomponent Isotherm model***

## INTRODUCTION

Urban solid waste includes house hold garbage and rubbish, street sweeping, construction and demolition debris, sanitation residues, trade and industrial refuse and bio-medical solid waste[1]. Solid waste management is the basic essential services provided by urban local bodies. However it is among the most poorly rendered service and the practices are unhygienic and inefficient.

Presently waste is littered everywhere and with rapid growth in population and urbanization the situation becomes critical[2]. The per capita waste generation in the country is around 1, 60,000 MT per day at a per capita waste generation ranging between 0.2 to 0.6kg per day. The waste generation is expected to increase at a rate of 1.33% per annum. With growth of urban population ranging between 3 to 3.5% per annum the annual increase in overall quantity of solid waste is assessed at about 5% and it translates to 260million tones per year by 2047[3].

About 90% of wastes are open dumped in landfills[1]. The major objectives of landfiling is to remove from general circulation the materials that are no longer in use, and to make the waste not to cause any impact on environment and on human health.

The unscientific disposal practices, i.e., the open dumping of waste cause severe impact on all the environmental attributes and on human health [4,5,6,7,8]. In India, MSW is disposed off in low-lying areas without any precautions or operational control. Hence the handling, management and disposal of municipal solid waste (MSW) throw a big challenge in all metropolitan cities[9].

The open dumped municipal solid waste under goes various phases of decomposition by microbial action. As the time progresses the open dumped solid waste undergo aerobic decomposition during the initial stages followed by onset of anaerobic conditions leading to dissolution of carbon dioxide and results in formation of acidic leachate and other organic acids. During the latter stage of anaerobic decomposition based on the availability of substrate, waste reduction is achieved by formation of methane[10].

The continuous deposition of land-filled solid waste, due of pressure will result in quizzing of a contaminated liquid as leachate. Leaching can be defined as the dissolution of a soluble constituent from a solid phase into a solvent. Leaching occurs as a consequence of the chemical reactions taking place at the scale of the individual waste particles as well as of the contaminant transport processes via the fluid moving through the solid particles. Many researchers has approximated that the quantity of leachate from uncovered or sparely vegetated MSW landfills lie between 15 to 60% of yearly precipitation[11,12,13,14].

Leachate contains dissolved, suspended and microbial contaminants from solid waste. Its high organic content, soluble salts and other constituents are capable of polluting ground water. The complex issue associated with the landfilling of the waste is its potential risk of leaching, particularly inorganic salts, heavy metals and toxic organic compounds[15].

The heavy metal concentration in preliminary phase of waste degradation may be higher and its presence in the leachate will vary widely. The low concentration of heavy metals in methanogenic leachates is not due to lack of heavy metals in the waste. Heavy metal balances for landfills have shown that less than 0.02% of heavy metals received at landfills are leached from the landfill after 30 years[16,17,18].

The mechanism controlling the leaching factor are therefore dependent on physical factors pertaining to both properties of waste material and the related fluid flow characteristics and also by the chemical factors like solubility of contaminants in waste. The extent of impact depends on the rate at which leaching occurs[15]. Generally the prevention of leachate migration in sanitary landfill will be achieved by natural or synthetic liner and a leachate collection system.

Fly ash is the inorganic pozzalonic residue obtained from lignite after combustion in boilers. It mainly consists of silica, alumina, and calcium oxide. The lignite and sub bituminous coal on combustion produce class C fly ash, while anthracite and bituminous coal under similar process generate Class F fly ash.

The sum of $SiO_2$, $Al_2O_3$ and $Fe_2O_3$ content in fly ash should be greater than 70% for class F fly ash whereas CaO content should be less than 5%. The class C fly ash are those fly ashes having less than 50% of $SiO_2$, $Al_2O_3$, $Fe_2O_3$ combined. CaO content varies from 20% to 30% for such fly ash. The lignite fly ash has predominantly quartz, anorthite, genlenite, hematite and mullite as major crystalline phases[19]. Table1 gives the characteristics of leachate used for the study. The typical characteristics of lignite fly ash are given Table 2.

Shaobin and Hongwei, (2006)[20] reviewed on Environmental-benign utilisation of fly ash as low-cost adsorbents and reports that the fly ash is a waste substance from thermal power plants, steel mills, etc. that is found in abundance in the world. In recent years, utilisation of fly ash has gained much attention in public and industry, which will help reduce the environmental burden and enhance economic benefit. Shaobin and Hongwei, (2006) [20] reported the technical feasibility of utilisation of fly ash as a low-cost adsorbent for various adsorption processes for removal of pollutants in air and water systems. Instead of using commercial activated carbon or zeolites, a lot of researchers have conducted the experiments using fly ash for adsorption of $NO_x$, $SO_x$, organic compounds, and mercury in air, and cations, anions, dyes and other organic matters in waters. It is recognised that fly ash is a promising adsorbent for removal of various pollutants. Chemical treatment of fly ash will make conversion of fly ash into a more efficient adsorbent for gas and water cleaning. Investigations also revealed that unburned carbon component in fly ash plays an important role in adsorption capacity[21].

Banerjee et.al., (1995)[22] reported that the fly ash is considered as a low cost sorbent and it is widely available. More over fly ash is an efficient sorbent, useful for water treatment involving removal of dissolved organic carbon. The fly ash is widely used as an additive in cement industry since it has Pozzolonic and cementitious properties. The lignite based coal fly ash used for this study has been obtained from the Neyveli Lignite Corporation, a Public sector thermal power plant located at a distance of 245km south of Chennai, the capital city of the state of Tamilnadu, India.

In this investigation an attempt is made to study the applicability of Fly ash, a Pozzolonic material for adsorption of multi heavy metal ions in leachate. Leachate samples were collected from the leachate pit, Mayiladuthurai Municipal solid waste open dumping site, Nagapattinam District, Tamil Nadu, India. Three Multi component adsorption isotherm models namely Extended Langmuir isotherm model, Langmuir-Freundlich isotherm model and Multicomponent isotherm model were used for modeling the equilibrium data of multi heavy metal ions such as Cu, Zn, Pb, Cr and Ni in leachate by Fly Ash as adsorbent.

## MATERIALS AND METHODS

The collected MSW leachate was analyzed and the contents are given in Table 1. All the analysis was carried out as per the procedure given in APHA 2005[23] at room temperature. The collected Fly Ash C was oven dried at 105ºC for a period of two hours**.** The oven dried adsorbent was under gone for the preliminary physicochemical analysis and tested for the presence of initial heavy metal concentration and the report is shown in Table 2. The heavy metal analysis in MSW leachate was carried out using Atomic Absorption Spectrophotometer (AAS- Make Thermo fisher - Chemito, Model 201). 100 ml of MSW leachate stock solution was taken in 250ml conical flask along with 0.2gms of fly ash and kept in rotary shaker for a period of maximum of 4hours in order to reach equilibrium. The shaker is maintained at constant rpm of 60. For a periodic time interval of 0, 30, 60, 90, 120, 150, 180, 210 and 240minutes the samples were collected. The samples were centrifuged and the supernatant were tested in AAS and the absorbances were noted at respective wave lengths. The concentration of multi heavy metal ions is calculated from the absorbance using the calibration curve. The amount of metal ions adsorbed on to the fly ash is calculated with mass balance equation 1.

$$q_{eq} = \frac{V}{M}\left(C_i - C_{eq}\right) ----(1)$$

Where qeq (mg/g) = uptake of metal ions at equilibrium conditions. Ceq (mg/l) = Concentration of metal ions at equilibrium condition. V (l) = Volume of solution taken for analysis. M (g) = Mass of adsorbent used. Ci (mg/l) = Initial concentration metal ion in MSW leachate. The same procedure is repeated with varying fly ash dosage of 0.4, 0.6, 0.8, 1.0 and 1.2g. All the experiments and analysis were carried out at a room temperature of 30ºC. The solution pH is maintained at 7 in all experiments.

The scanning electron microscopy (SEM) is widely used to study the formation and morphology of adsorbent. The major advantage of SEM is its ability to study the heterogeneity of materials to visualize various mineral components in their distinct growth forms and their relation in terms of overall micro fabric and texture. The SEM micro graph of the fly ash show spherical morphology of the fly ash particles. It is constituted by compact spheres of different sizes; on the sphere surface the existence of solid deposits or small crystals is also observed. The fly ash is mostly free on the surface. The size varying between 1um to 10um. The SEM analysis was performed both before and after adsorption and the pictures were given in Figure 1 and Figure 2.

## RESULTS AND DISCUSSION

*Adsorption Isotherms*

The equilibrium relationships between adsorbent and adsorbate are resulted by the study of adsorption isotherms which are usually the ratio between the quantity adsorbed and the remaining in solution at fixed temperature at equilibrium. Also the capacity of the adsorbent is calculated by the equilibrium studies. Adsorption Equilibria provides the fundamental physicochemical information to assess the applicability of the adsorption process. Moreover the Adsorption Equilibria is the essential to analysis and design of the adsorption process. It describes the phenomenon governing the mobility of a substance from the aquatic environments to a solid-phase under equilibrium condition at a constant temperature and pH[24,25,26].

The equilibrium adsorption isotherm is very important to design the adsorption systems. Adsorption equilibrium is established when an adsorbate containing phase has been contacted with the adsorbent for sufficient time, with its adsorbate concentration in the bulk solution is in a dynamic balance with the interface concentration[24,27,28]. The mathematical correlation, which provides an important role towards the modeling, operational design and applicability of the adsorption systems, is usually described by graphically and expression of adsorbate concentration in the solid-phase against liquid phase[24,29].

` The nature of interaction between adsorbate and adsorbent at the time of equilibrium could be determined by the correlation of experimental data with equilibrium isotherms. Various single component isotherms like Langmuir, Freundlich, Dubinin-Radushkevich, Temkin, Toth, Redlich-Peterson, Sips, Khan and Hill models have been studied but unexpectedly no one would be a successful, due to the presence of multi heavy metals in the leachate used in this investigation. This unfit nature is the account of presence of other metal ions which may either interact with adsorbate of interest or compete for same binding sites of adsorbent. Due to these constraints, the amount of adsorbent adsorbed either increase, decrease or remain unaltered in the presence of other heavy metal ions. A number of alternates have been proposed to evaluate the multicomponent adsorption. In the present investigation three multi component adsorption isotherms namely Extended Langmuir isotherm, Langmuir-Freundlich isotherm and Multicomponent Adsorption Isotherm has been used to assess adsorbate -adsorbent interactions and nature of adsorption[30]. Table 3 shows the equations and parameters of multicomponent isotherms investigated in this present study.

*Extended Langmuir Isotherm*

When two or more adsorbable components exist with the possibility of occupying the same adsorption sites, isotherm relationship become more complex. The simplest is the extension of Langmuir type isotherm by assuming no interaction between adsorbate[31]. Commercial application of physical adsorption involves mixture rather than pure components. During the adsorption of a component from the liquid solution containing n number of components, certainly one can increase, decrease or have no influence on the adsorption of other component, depending upon the interactions of the adsorbed molecules. Mohan and Chander, (2001)[32]; Aksu and Gilen, (2002)[33] and Bhumica and Chandrajit, (2013)[34] reports the three effects of competitive adsorption obviously. Presence of two or more than two components in aqueous systems may have three types of effects: synergism - that is the effect of the mixture is greater than that of each of the individual effects of the constituents in the mixture, antagonism - that is the effect of mixture is less than that of the each of the individual effects of the constituents in the mixture and non-interaction - that is the effect of the mixture is neither more nor less than that of each of the individual effects of the constituents in the mixture. Many models have been published in the literature to describe the equilibrium relationship between adsorbate and adsorbent for single component system but unfortunately very least for multicomponent system. One of the most important model is Extended Langmuir Adsorption model which has permitted the calculation of $q_{eq,i}$ the amount of adsorbate i adsorbed per unit weight of adsorbent in the presence of another adsorbing adsorbate. The simple theoretical treatments to the extension of the Langmuir equation by neglecting such an interactions and assumes that the only effect is the reduction of vacant surface area to the adsorption. The extended Langmuir isotherm model was applied to determine the adsorption of multi heavy metal ions in leachate onto the Fly Ash adsorbent.

The extended Langmuir isotherm model is shown in Table 3. The extended Langmuir parameters $k_i$ and $b_i$ can be obtained by using the fminsearch tool in Matlab10a Version. Table 4 shows the constants and parameters of Extended Langmuir isotherm for adsorption of Multi heavy metal ions in leachate by Fly Ash solid adsorbent. The coefficient of correlation for Extended Langmuir isotherm is found to be good. Figure 3 shows the plot $q_{eq}$ experimental against $q_{eq}$ by extended Langmuir isotherm. The coefficient correlation $R^2$ indicates that the experimental equilibrium uptake and calculated equilibrium uptake concur with one another. Figure 3 ensures the same. Hence the Extended-Langmuir Adsorption isotherm is found to be the best fit for adsorption of multi heavy metal ions in MSW leachate by Lignite based coal fly ash. To evaluate the interaction effect of more than two adsorbents, the ratio of adsorption capacity of one adsorbate ($q_{mix}$) in the MSW leachate to that of same adsorbate when present alone ($q_o$) was calculated for the following conditions as reported by Mohan and Chander., (2001)[32] and Bhumica and

Chandrajit.,(2013) [34] i.e., qmix/qo > 1, synergism; qmix/qo = 0, non-interaction and qmix/q0 < 1, antagonism.

For all the heavy metals, considered in this study, the value of qmix/qo is greater than zero indicating that the non-interaction between the adsorbent and adsorbate never occurred and a healthy competition between the metal ions present in the MSW leachate exists, due to the presence of more active sites. This is further confirmed by the values obtained for qmix/qo in which synergism is followed by antagonism. Table 5 shows the qmix/qo values for Extended Langmuir adsorption isotherm. It is also observed in Table 5 that the Nickel ion shows a higher synergism to adsorb on to the fly ash for all values of adsorbent dosage and it is followed by Chromium up to a dosage of 0.8 gms of fly ash. Up to a dosage of 0.2 gms of fly ash, Pb and Zn ions shows the synergism since qmix/q0 is greater than 1, later antagonism is observed for both metal ions. For Cu, synergism exists up to an adsorbent dosage of 0.4 gms of fly ash later it turns out to be antagonism.

*Langmuir-Freundlich Isotherm*

A combination of Langmuir and Freundlich isotherm models makes a new model called the Langmuir-Freundlich isotherms model. The Langmuir Freundlich isotherm model parameters Ki, bi and ni are obtained by using the fminsearch tool in Matlab10a Version. Table 6 shows the constants and parameters of Langmuir Freundlich adsorption isotherm. The coefficient of correlation obtained for the comparison of qeq experimental with qeq Langmuir Freundlich isotherm is found to be least for all heavy metal ions present in leachate. It indicates that Langmuir-Freundlich Adsorption isotherm model for competitive adsorption of metal ions has least significance since the $R^2$ values are less. Figure 4 shows the graphical representation for the plot of $q_{eq}$ experimental with $q_{eq}$ Langmuir - Freundlich isotherm.

*Multicomponent Isotherm*

The equation for Multicomponent adsorption isotherm is given in Table 3. The constants and parameters are given in Table 7 for Multicomponent Adsorption isotherm. Figure 5 shows the graphical fit of $q_{eq}$ experimental with $q_{eq}$ of Multicomponent isotherm. The coefficient of correlation for Multicomponent adsorption isotherm is found to be least for all heavy metal ions present in the leachate when compared to Extended Langmuir Model for the adsorption of multiheavy metal ions present in leachate by Fly Ash as adsorbent.

## CONCLUSIONS

The present studies show the applicability of Fly Ash, the pozalonic material for the adsorption of multi heavy metal ions in MSW leachate. Among the three isotherms Extended Langmuir adsorption isotherm model shows a greater fit, rather than Langmuir-Freundlich Adsorption isotherm model and Multicomponent adsorption model. The coefficient of correlation $R^2$ found to be high for all the metal ions by Extended Langmuir adsorption isotherm model.

For all the heavy metals, considered in this study, the value of qmix/qo is greater than zero indicating that the non-interaction between the adsorbent and adsorbate never occurred and a healthy competition between the metal ions present in the MSW leachate exists, due to the presence of more active sites, this is further confirmed by the values obtained for qmix/qo in which synergism is followed by antagonism. Table 5 shows the qmix/qo values for Extended Langmuir adsorption isotherm. It is also observed in Table 5 that the Nickel ion shows a higher synergism to adsorb on to the fly ash for all values of adsorbent dosage and it is followed by Chromium up to a dosage of 0.8 gms of fly ash. Up to a dosage of 0.2 gms of fly ash, Pb and Zn ions shows the synergism since qmix/q0 is greater than 1, later antagonism is observed for both metal ions. For Cu, synergism exists up to an adsorbent dosage of 0.4 gms of fly ash later it turns out to be antagonism.

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Table 1: Properties of MSW Leachate

| Constituent in MSW Leachate | Concentration |
|---|---|
| pH | 7.61 |
| EC | 5.64 |
| Chlorides | 1640 |
| Hardness | 1040 |
| Turbidity | 47 |
| Sulphate | 295 |
| Fe | 27.96 |
| Ammonia | 50 |
| Na | 98 |
| Ca | 282 |
| K | 45 |
| Phosphate | 14 |
| COD | 3920 |
| Acidity | 70 |
| BOD | 1224 |
| Cu | 1.22 |
| Zn | 2.86 |
| Pb | 2.62 |
| Cr | 1.5 |
| Ni | 0.65 |
| Cd | BDL |

All values are in mg/l except pH and EC

Table 2: Characteristics of Fly ash used for this study

| Sl.no | Parameter | Percentage |
|---|---|---|
| 1. | Silica as $SiO_2$ | 62.6 |
| 2. | Iron as $Fe_2O_3$ | 4.04 |
| 3. | Alumina as $Al_2O_3$ | 24.41 |
| 4. | Calcium as CaO | 0.35 |
| 5. | Magnesium as MgO | 0.54 |
| 6. | Sulphate as $SO_3$ | 0.85 |
| 7. | Loss on Ignition | 1.27 |
| 8. | pH | 8.12 |
| 9. | EC | 0.44 |
| 10. | TDS (mg/l) | 286 |
| 11. | Chlorides (mg/l) | 24 |
| 12. | Hardness (mg/l) | 2250 |
| 13. | Ca (mg/l) | 198.50 |
| 14. | Na (mg/l) | 41.55 |
| 15. | K (mg/l) | 6.15 |
| 16 | $SO_4$ (mg/l) | 108 |
| 17. | p (mg/l) | 0.0 |
| 18. | Fe (mg/l) | BDL |
| 19. | Cd (mg/l) | 0.04 |
| 20. | Cu (mg/l) | 0.03 |
| 21. | Zn (mg/l) | BDL |
| 22 | Pb (mg/l) | 0.03 |

Table 3: Equations and parameters of Multicomponent isotherm

| Model | Equation | Parameters |
|---|---|---|
| Extended-Langmuir Isotherm Model[25, 25a 25b 25c 25d] | $q_{eq,i} = \frac{k_i C_{eq,i}}{1+\sum_{i=1}^{n} b_i C_{eq,i}} --(1)$ | k and b |
| Langmuir-Freundlich Isotherm[26,27] | $q_{eq,i} = \frac{k_i C_{eq,i}^{n,i}}{1+\sum_{i=1}^{n} b_i C_{eq,i}^{n,i}} --(2)$ | k, b and n |
| Multicomponent Isotherm[26,28] | $q_{eq,i} = \frac{k_i C_{eq,i}}{1+\sum_{i=1}^{n} b_1 C_{eq,i}^{n,i}} --(3)$ | k, b and n |

Table 4: Extended Langmuir Adsorption isotherm for adsorption of Multi heavy metals in leachate by Fly Ash solid adsorbent

| Heavy Metal | Extended Langmuir Adsorption Isotherm Parameters and Constants | | |
|---|---|---|---|
| | K | b | $R^2$ |
| Copper Cu | 0.4479 | 0.4007 | 0.899 |
| Zinc Zn | 0.1359 | 0.4219 | 0.902 |
| Lead Pb | 0.1440 | 2.3127 | 0.857 |
| Chromium Cr | 0.2073 | 2.0652 | 0.923 |
| Nickel Ni | 0.5820 | 0.3588 | 0.897 |

Table 5: Calculated values of $q_{mix}/q_o$ for Extended Langmuir Adsorption Model.

| Adsorbent dosage | Cu $q_{mix}/q_o$ | Zn $q_{mix}/q_o$ | Pb $q_{mix}/q_o$ | Cr $q_{mix}/q_o$ | Ni $q_{mix}/q_o$ |
|---|---|---|---|---|---|
| 0.2 | 8.684 | 1.001 | 7.101 | 18.25 | 8.5935 |
| 0.4 | 1.495 | 0.181 | 1.001 | 2.77 | 1.7774 |
| 0.6 | 0.945 | 0.124 | 0.597 | 1.70 | 1.3601 |
| 0.8 | 0.468 | 0.080 | 0.305 | 1.00 | 1.0853 |
| 1.0 | 0.265 | 0.032 | 0.175 | 0.65 | 0.9979 |
| 1.2 | 0.150 | 0.038 | 0.086 | 0.37 | 1.1694 |

Table 6: Langmuir-Freundlich Adsorption isotherm for adsorption of Multi heavy metals in leachate by Fly Ash solid adsorbent

| Heavy Metal | Langmuir-Freundlich Adsorption Isotherm Parameters and Constants | | | |
|---|---|---|---|---|
| | K | b | n | $R^2$ |
| Copper Cu | 1.3745 | 3.0532 | 0.2364 | 0.806 |
| Zinc Zn | 6.2283 | 2.4680 | 0.5363 | 0.064 |
| Lead Pb | 2.8558 | 3.6312 | 1.1248 | 0.776 |
| Chromium Cr | 1.0356 | 2.1711 | 0.3991 | 0.805 |
| Nickel Ni | 1.4334 | 1.4195 | 0.5511 | 0.548 |

Table 7: Multicomponent Adsorption isotherm for adsorption of Multi heavy metals in leachate by Fly Ash solid adsorbent

| Heavy Metal | Multicomponent Adsorption Isotherm Parameters and Constants | | | |
|---|---|---|---|---|
| | K | b | n | $R^2$ |
| Copper Cu | 1.3831 | 15.4120 | 1.5957 | 0.861 |
| Zinc Zn | 0.2675 | 45.4468 | 2.2427 | 0.867 |
| Lead Pb | 0.6721 | 2.5651 | 0.6298 | 0.816 |
| Chromium Cr | 0.3768 | 8.7826 | 5.3524 | 0.887 |
| Nickel Ni | 0.1528 | 2.0224 | 0.2209 | 0.865 |

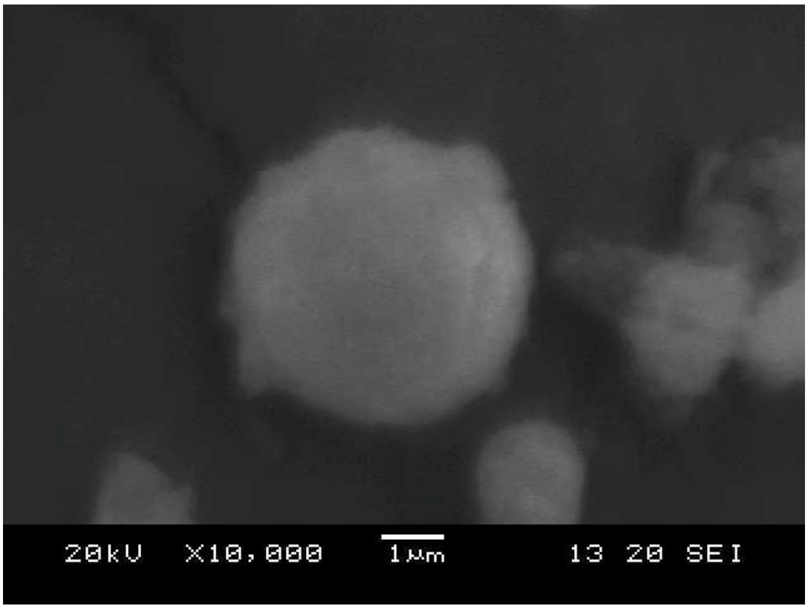


**Figure 1SEM image of Individual fly ash before adsorption**

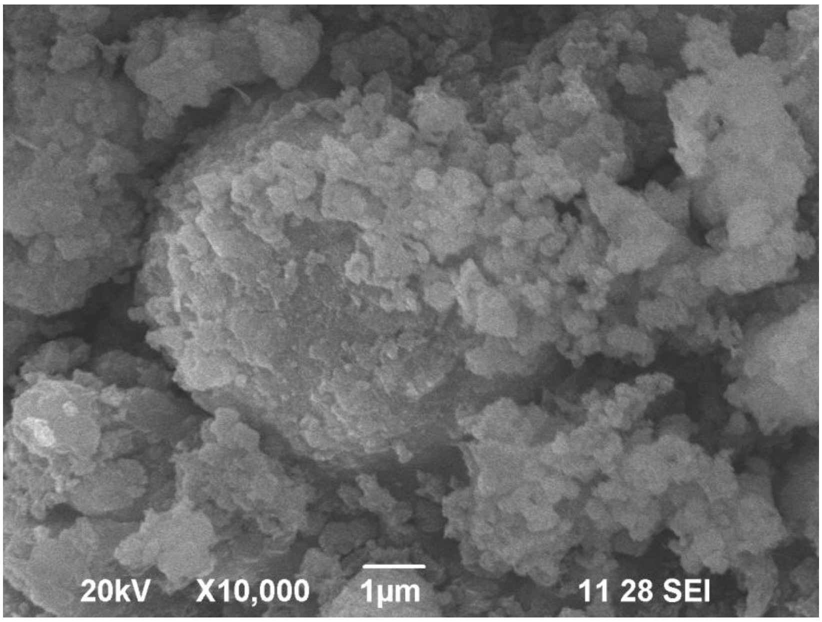


Figure 2 SEM image of fly ash after adsorption

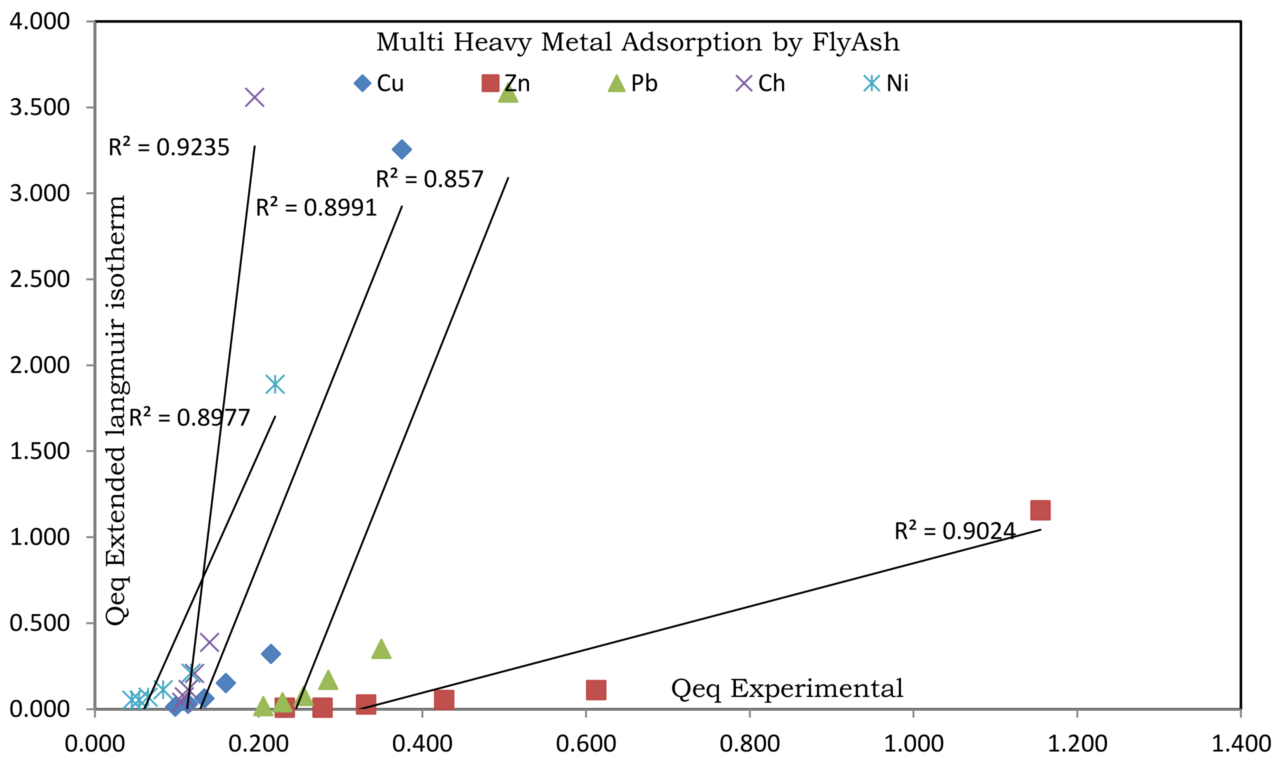


Figure 3: Extended Langmuir Adsorption isotherm for adsorption of Multi heavy metals in leachate by Fly Ash solid adsorbent

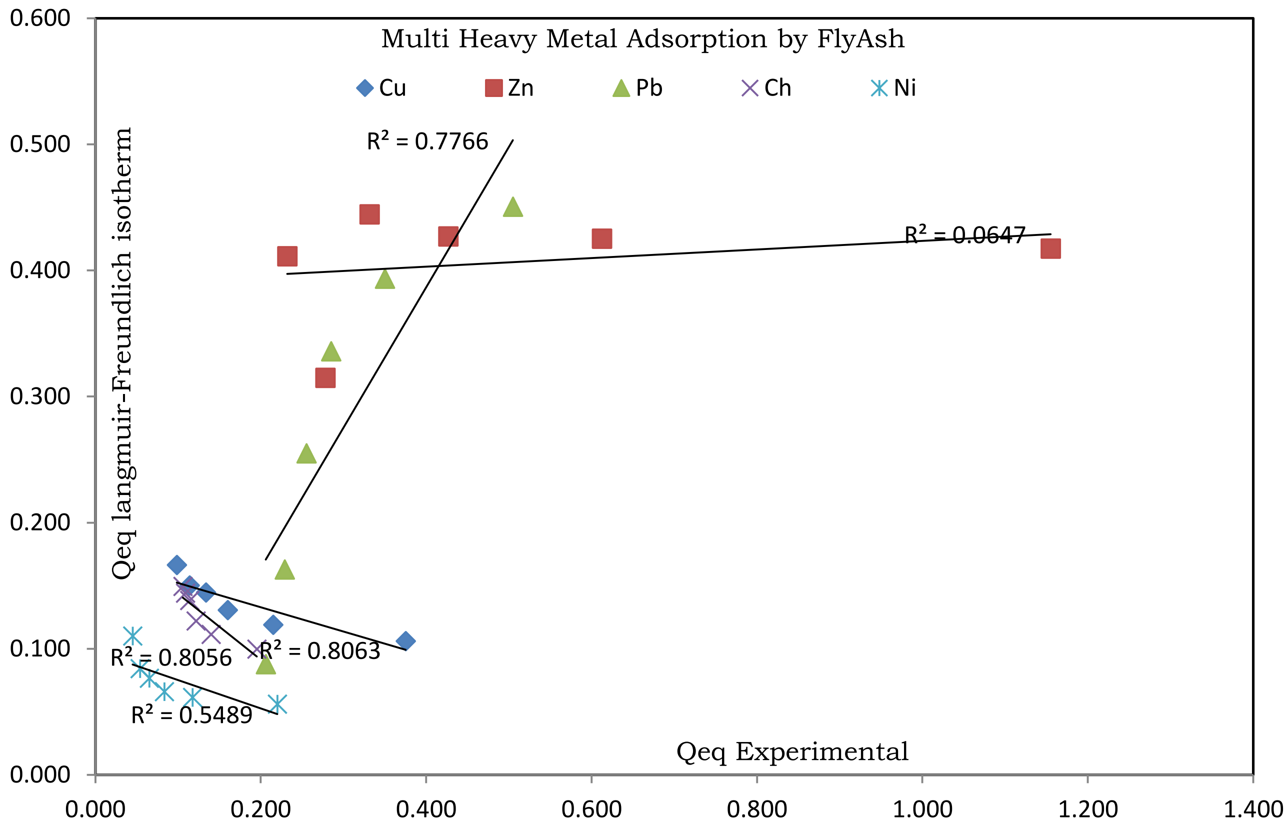


Figure 4: Langmuir-Freundlich Adsorption isotherm for adsorption of Multi heavy metals in leachate by Fly Ash solid adsorbent

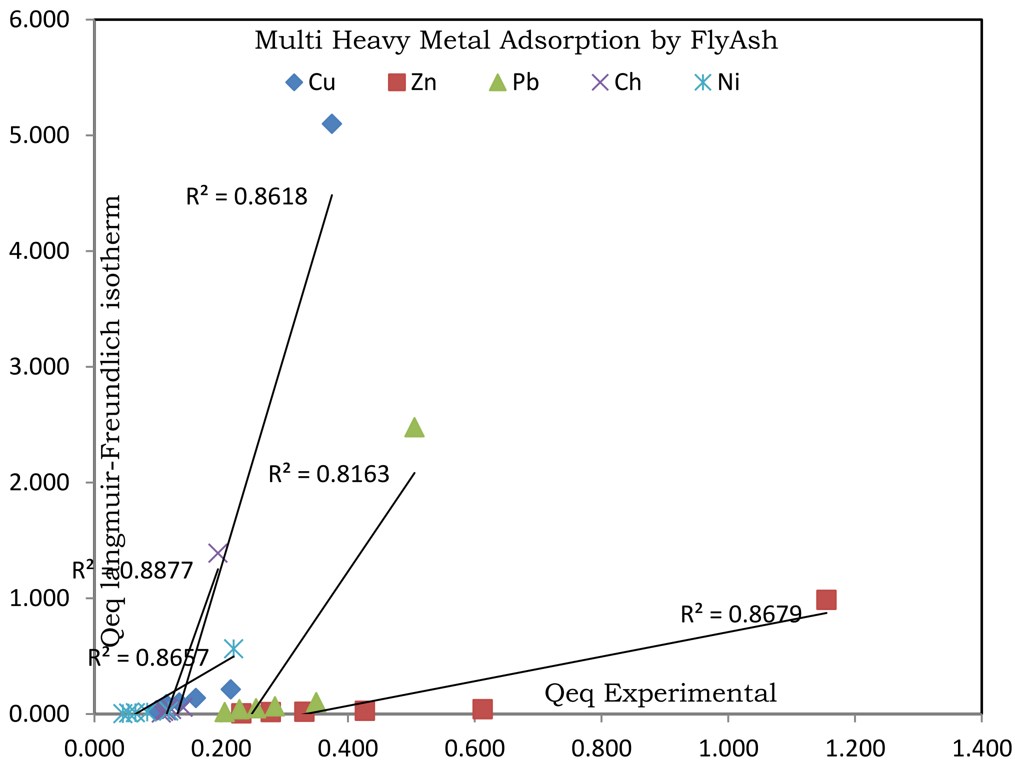


Figure 5: Multicomponent Adsorption isotherm for adsorption of Multi heavy metals in leachate by Fly Ash solid adsorbent